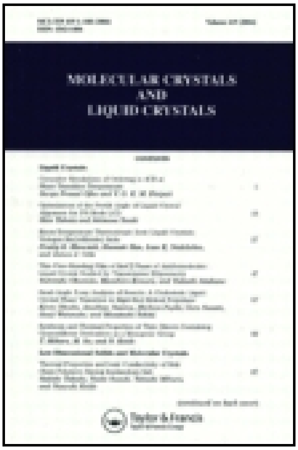

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Dielectric Response of Ferroelectric Liquid Crystals in Helical and Twisted Planar Samples

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Dielectric Response of Ferroelectric Liquid Crystals in Helical and Twisted Planar Samples

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The dielectric spectroscopy has been studied for both twisted and helical structures at the same sample. On cooling below the SmA phase the twisted structure spontaneously appeared. The helical structure could be generated by applying electric field of the frequency ~20 Hz. It is shown that the twisted structure exhibits higher relaxation frequency and lower dielectric strength than the helical one. This result can be explained within the present theory providing the existence of weak depolarization fields.

Keywords: ferroelectric liquid crystals; dielectric spectroscopy; helical structure; twisted structure

INTRODUCTION

In planar samples of the ferroelectric SmC* phase either the helical structure accompanied with dechiralization lines or structure with unwound helix can appear. The unwound sample can be completely homogeneous, or possesses a twist within the smectic layers fixed by the polar surface anchoring. The described structures take place according to the sample thickness and sample processing. Dielectric properties of such structures that occur in real sample cannot be described just by the Goldstone and soft mode as in an ideal unbound SmC* structure.

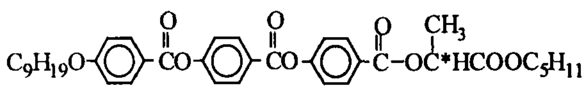
The dielectric response of described structures (homogeneous, twisted and helical with the dechiralization lines) has been studied experimentally and has been found strongly dependent on the sample thickness. Theories worked out to explain the dielectric properties of the homogeneous [1] and twisted structure [2] confirm thickness dependences of both relaxation frequency and dielectric strength.

In the present contribution a theory treating the dielectric response of the helical ferroelectric structure confined in a planar sample is presented. It is supposed that the helix is pinned by depolarization lines, which exist in the sample due to the homogeneous surface anchoring. The theory yields thickness dependent dielectric response. The experimental results obtained with two ferroelectric liquid crystals for both twisted and helical sample structure are mutually compared and explained on the basis of described models.

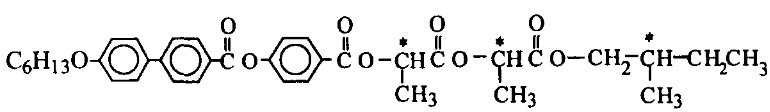
EXPERIMENTAL

We studied substances denoted as 9AL (synthesis and basic properties described in [3]) and ZLL6/*:

9AL: Cr - 48°C - SmC* - 80°C - SmA - 129°C - Iso



ZLL6/*: Cr - 66°C - SmI* - 76°C - SmC* - 97.5°C - SmA - 142°C - Iso



The cells were filled in the isotropic phase and composed of glass plates provided with ITO electrodes and polyimide layers unidirectionally rubbed, which ensures planar (bookshelf geometry). The sample thickness was defined by mylar sheets (6, 12, 25, 50, and 100 μm). To get a good alignment, the samples were slowly cooled (0.1K/min.) from the isotropic to the SmA phase. The sample texture was checked in a polarizing microscope. On cooling from the SmA phase the helical structure with characteristic dechiralization lines appears in samples thicker than 6 μm for 9AL and 12 μm for ZLL6/*. In thinner samples the helix is spontaneously unwound, but a director twist-bend appears along the sample normal (twisted structure). In twisted samples the helix can be generated by application of a weak a.c. field (typically 5×10^5 V/m, 20 Hz).

Dielectric dispersion was measured in the frequency range from 5 Hz to 1 MHz by the Solartron impedance analyzer at stabilized temperatures.

EXPERIMENTAL RESULTS

The dielectric response exhibits one relaxation process in the whole studied temperature region for both studied materials. The relaxation frequency f_r and dielectric strength $\Delta\varepsilon$ are determined by fitting the measured data to Cole-Cole formula.

In Figure 1 there is a temperature dependence of f_r for ZLL6/*

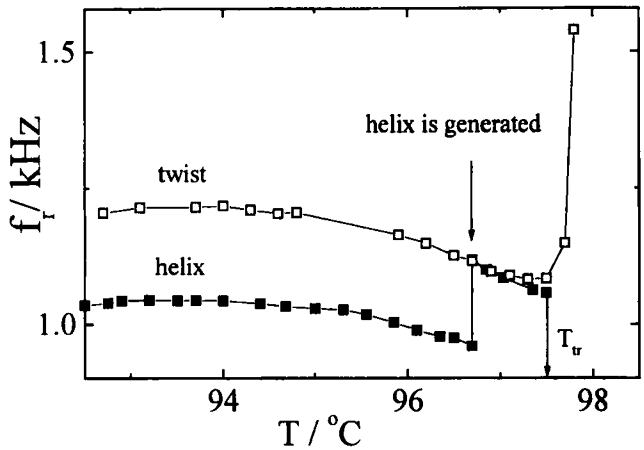


FIGURE 1 Temperature dependence of the relaxation frequency for ZLL6/* sample 12 μm thick for twisted and helical structures.

measured on the sample 12 μm thick for both twisted and helical structure. The twisted structure appeared on cooling from the SmA phase and persisted down to about 90°C; on continuing cooling the helix gradually appears. In the next cooling run the helical structure was generated by the electric field near below the SmA-SmC* phase transition temperature T_{tr} and persisted even if the field is switched of.

The change of f_r and $\Delta\varepsilon$, when the structure is transformed from the twisted to the helical one, is seen in Figure 2 for thin samples of the studied materials. On cooling, just below the temperature T_{tr} the values correspond to the twisted structure. At a temperature in the SmC* phase the helical structure is generated by application the a.c. electric field. For both materials the change of the structure is manifested by a jump down in f_r and jump up in $\Delta\varepsilon$ on cooling.

The results for relaxation frequency as well as for the dielectric strength exhibit strong thickness dependence in both twisted and helical structures similarly as in [1,2,4].

THEORY

The dynamic dielectric response of a liquid crystal layer of a finite thickness in planar geometry can be determined from the relaxation equation [2,5] constructed on the basis of the free energy

$$\gamma\theta_S^2\frac{\partial\phi}{\partial t}=\theta_S^2\left(K_1\frac{\partial^2\phi}{\partial x^2}+K_3\frac{\partial^2\phi}{\partial z^2}\right)-P_SE_x(t)\sin\left(\phi+\frac{\pi}{2}\right) \qquad (1)$$

The macroscopic electric field $E_x(t)= E_x(t)+ E_{dep,x}(t)$ involves both the external electric field $E_x(t)$ and the depolarization field $E_{dep,x}(t)$. It was assumed that the spontaneous tilt angle θ_S is time and spatially independent, γ is the rotational viscosity, the elastic constants are K_1 in smectic layer, and K_3 perpendicular to smectic layer. Inserting the homogeneous a.c. electric field $E_x(t)= E\,e^{i\omega t}$ the equilibrium (*twisted* or *helical*) structure described by the spontaneous azimuthal angle $\phi_S(x,z)$ is perturbed, so that

$$\phi(x,z,t) = \phi_S(x,z) + \delta(x,z)\,e^{i\omega t}, \qquad (2)$$

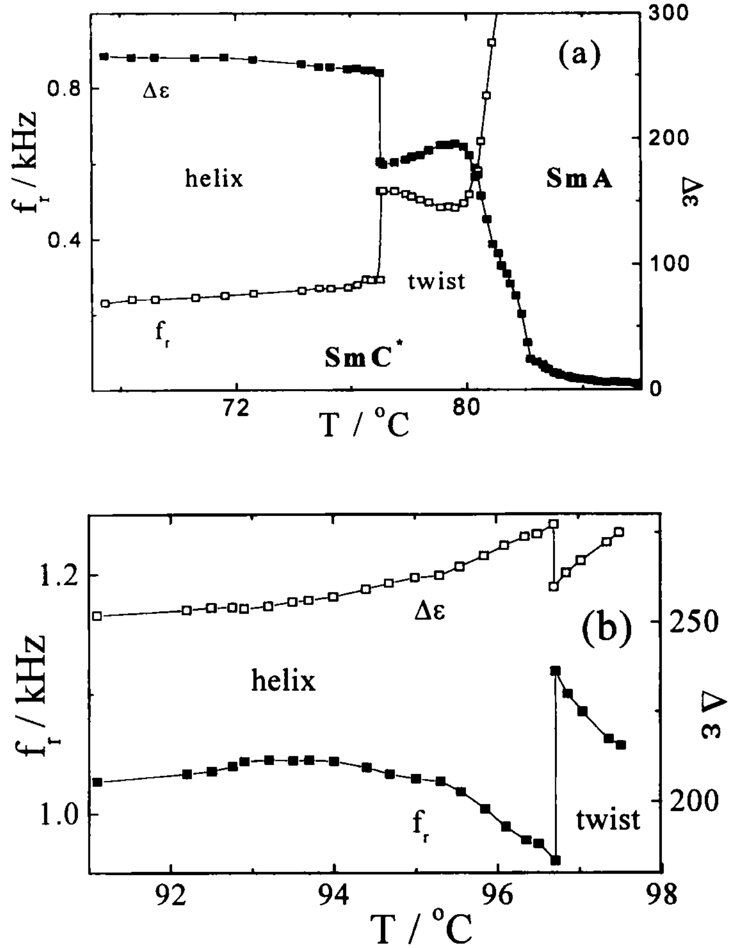


FIGURE 2 Temperature dependence of the relaxation frequency and the dielectric strength for (a) 9AL, 6 μm thick, in both SmA and SmC* phases and (b) ZLL6/*, 12 μm thick, in the SmC* phase only. The jumps in the SmC* phase corresponds to the change of the structure from twisted to helical one.

where δ(x,z) is a small perturbation independent of the y-axis. The pinning of the structure at the surfaces is expressed by the boundary conditions:

$$\delta(x=0,z)=\delta(x=d,z)=0\,. \tag{3}$$

The polarization deviates from its equilibrium value by (δP_x, δP_y, 0), where $\delta P_x = -P_S\cos(\phi(x,z))\delta(x,z)$ and $\delta P_y = -P_S\sin(\phi(x,z))\delta(x,z)$.

If the time evolution of polarization is faster than the screening process the depolarization field appears

$$\operatorname{div}(\varepsilon_0\mathbf{E}_{dep}) = -L\operatorname{div}\delta\mathbf{P} = -L\partial\delta P_x/\partial x\,, \tag{4}$$

where L is a depolarization factor smaller than 1 due to the partial charge compensation.

The schemes of the twisted and helical structures are shown in Figs. 3 a and b, respectively.

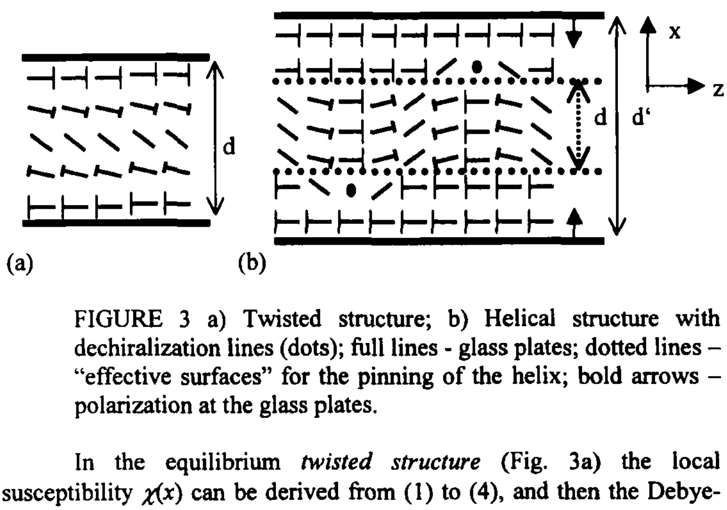


FIGURE 3 a) Twisted structure; b) Helical structure with dechiralization lines (dots); full lines - glass plates; dotted lines – “effective surfaces” for the pinning of the helix; bold arrows – polarization at the glass plates.

In the equilibrium *twisted structure* (Fig. 3a) the local susceptibility $\chi(x)$ can be derived from (1) to (4), and then the Debye-like total susceptibility is obtained with the static value χ_0 and the relaxation frequency $f_r = (2\pi\tau)^{-1}$:

$$\chi_0 = \frac{4\pi\mu^2/2}{K_1(\pi/d)^2 + L_t\mu^2/2\varepsilon_0} \quad \text{and} \quad f_t = (2\pi\tau)^{-1} = \frac{1}{\gamma}\left(\frac{K_1\pi^2}{d^2} + \frac{L_t\mu^2}{2\varepsilon_0}\right), \quad (5)$$

where $\mu \equiv P_S/\theta_S$, depolarization factor of twisted structure is denoted L_t.

The equilibrium ***helical structure*** is pinned at the "effective surfaces" (Fig. 3b) and it depends on the z-axis only; the response of the surface layers of the thickness *(d'-d)/2* being neglected. Eqs. (1) to (4) can be approximately solved and the total susceptibility is obtained in the form:

$$\chi = \left(1 - \frac{\tanh\left(\sqrt{(i\gamma\omega + K_3q^2 + L_h\mu^2/2\varepsilon_0)/K_1}\,\frac{d}{2}\right)}{\sqrt{(i\gamma\omega + K_3q^2 + L_h\mu^2/2\varepsilon_0)/K_1}\,\frac{d}{2}}\right)\frac{4\pi\mu^2/2}{i\gamma\omega + K_3q^2 + L_h\mu^2/2\varepsilon_0} \quad (6)$$

where L_h expresses depolarization factor of the helical structure.

DISCUSSION

For discussion typical experimental values are considered: $\theta_S = 15°$, $p = 1$ μm, $K_3 = 5\times10^{-12}$ N, $P_S = 50$ nC/cm^2. Strong anisotropy of the smectic structure implies $K_3 << K_1$, and we consider $K_1 = 50\times K_3$.

The main conclusions derived from (5) and (6) are:

(i) Response of both the twisted and helical structures exhibits Debye-like character. A nearly Debye-like response for the helical structure is confirmed by numerical inspection of (6), though generally this response is multi-relaxational.

(ii) With the increase of the sample thickness the permittivity of both structures increases. Non-zero depolarization factors L_t and L_h ensure that this dependence tends to saturation.

(iii) A reasonable agreement between the permittivity values measured with twisted sample 6μm thick and the result given by the theory is obtained if $L_t \approx 0.01 << 1$, which means that only 0.01 of the internal field remains non-compensated. For the helical structure a similar comparison yields $L_h < 0.001$.

(iv) Experimentally observed higher relaxation frequency of the twisted structure as compared with that of the helical structure requires $L_t > L_h$

in the model expressions (5) and (6). This relation was checked numerically [5] since the analytical form of the relaxation frequency of the helical structure is not available. This relation qualitatively agrees with estimation of depolarization factors obtained in (iii) from comparison of permittivities of both structures.

It can be concluded that the presented theory describes reasonably the dielectric response of finite SmC* samples exhibiting helical or twisted structure. The results derived for both relaxation frequency and dielectric strength are very sensitive to depolarization fields. A comparison with experiment shows that these fields must be almost compensated by internal charges, but not completely.

Acknowledgments

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